Comparative Assessment of Closures for Turbulent Reacting Flows

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In a recent article, Dutta and Tarbell (1989) made a comparative study of several turbulence closures for predicting the mean rate of reactant conversion in a chemical reaction of the type $A+B\rightarrow$products in homogeneous turbulent flows. In this work, we use recent developments in the approach based on the probability density function (PDF) method for treating the problem considered by Dutta and Tarbell (1989). With this method, we propose a simple algebraic relation for predicting the limiting rate of mean reactant conversion. The PDF model is based on the amplitude mapping closure (AMC) of Kraichnan (Kraichnan, 1989; Chen et al., 1989), whose superiority has been demonstrated in a number of validation studies (Pope, 1991; Gao, 1991; Frankel et al., 1992, Chen et al., 1989; Madnia et al., 1991b; Madnia et al., 1992). In accordance with the problem discussed by Dutta and Tarbell (1989), we consider a stoichiometric mixture with initially segregated reactants. This is convenient for expressing the final results in a simple algebraic form. However, it is understood that the AMC can be employed for modeling of nonequilibrium chemically reacting flows under arbitrary initial conditions (Pope, 1991).

For comparison, in addition to the closure of Toor (1962) and the three-environment (3E) model of Dutta and Tarbell (1989), several other closures are also considered. These are the PDF methods based on the generalized coalescence/dispersion (C/D) models and those based on "assumed" frequencies. For the C/D models, the closures of Curl (1963), and Janicka et al. (1979) are examined. The assumed distributions are based on the beta density of the first kind (Pearson, 1895) and the Logit-normal density (Johnson, 1949a). The applicability of these assumed densities for the problem under consideration has been ascertained by Madnia et al. (1991a) and Miller et al. (1993). Finally, to assess the performance of the models, the results predicted by all the closures are compared with those generated by direct numerical simulations (DNS). These simulations have proven very effective in validation studies of turbulent reacting flows (Eswaran and Pope, 1988; Givi and McMurtry, 1988; Givi, 1989; Leonard and Hill, 1991) and provide a useful tool in model assessments of the type pursued here.

Formulations and Model Presentation

With the assumption of an infinitely fast chemistry, all the pertinent statistics of the reacting field are related to those of an appropriate conserved scalar variable, hereby denoted by $\mathcal{J}$ (Hawthorne et al., 1949; Bilger, 1980). Under this assumption, in a stoichiometric mixture, the single-point PDF of the reacting scalar is related to that of the conserved scalar by (Bilger, 1980; Kosaly and Givi, 1987):

$$\mathcal{P}_A(\psi,t)=\frac{1}{2}\,\mathcal{P}_{\mathcal{J}}\left(\frac{\psi+1}{2},t\right)+\frac{1}{2}\,\delta(\psi),\quad 1>\psi>0. \tag{1}$$

Here, δ denotes the delta function. Therefore, based on Eq. 1, if the PDF of the conserved scalar is known, all the statistical information regarding the reacting scalars A and B is available.

C/D Closures

The C/D models consist of PDF transport equations, in which the effects of molecular mixing are modeled in terms of a particle-pair interaction process. The most general form of the model can be expressed by the evolution equation (Pope, 1982; Kosaly and Givi, 1987; Dutta and Tarbell, 1989):

$$\frac{\partial\mathcal{P}(\psi,t)}{\partial t}=-2\beta\omega\mathcal{P}(\psi,t)+2\beta\omega\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}d\psi'\,d\psi''\,\mathcal{P}(\psi',t)\mathcal{P}(\psi'',t)\times\int_{-\infty}^{\infty}d\alpha\mathfrak{A}(\alpha)\delta\left[\psi-(1-\alpha)\psi'-\frac{1}{2}\,\alpha(\psi'+\psi'')\right]. \tag{2}$$

In this equation, the random variable $\alpha\in[0,1]$ and the kernel $\mathfrak{A}(\alpha)$ controls the extent of mixing. The parameter β is the controlling factor which yields the same variance for all members of the C/D family and ω is the mixing frequency (Pope, 1982). Different C/D closures are obtained by different choices for the function $\mathfrak{A}(\alpha)$. This function is nonzero, nonnegative and normalized to unity within $\alpha\in[0,1]$. Some of the more widely utilized members of the C/D family are the models of

Curl (1963) and Janicka et al. (1979), and the LMSE closure of O'Brien (1980). These closures imply, respectively, $\mathfrak{C}(\alpha)=\delta(\alpha-1)$, $\mathfrak{C}(\alpha)=1$, and $\mathfrak{C}(\alpha)=\mathrm{Lim}_{\epsilon\to 0}\delta(\alpha-\epsilon)$. For the problem under consideration, the results obtained from the LMSE model are identical to those based on Toor's hypothesis (Kosaly and Givi, 1987).

With a combination of Eqs. 1 and 2, the mean reactant conversion rate is determined from the knowledge of $\mathcal{P}_A(\psi)$ [or $\mathcal{P}_B(\psi)$]. The numerical solution of the C/D transport equation (Eq. 2) can be obtained by Monte-Carlo methods (Pope, 1981).

Mapping Closure

The amplitude mapping closure (AMC) involves the mapping of the scalar field of interest to a Gaussian reference field. The knowledge of this mapping allows determination of the scalar PDF (Chen et al., 1989; Pope, 1991). For the binary problem under consideration here, the solution for the mapping function has been obtained by Pope (1991) and the resulting PDF is of the form:

$$\mathcal{P}(\psi)=\frac{\gamma}{\sqrt{2}}\exp\left\{-\left[\frac{\gamma^2}{2}-1\right][\mathrm{erf}^{-1}(2\psi-1)]^2\right\}, \tag{3}$$

where the parameter γ depends on the missing two-point information and is not known *a priori* in the context of a single-point description. It is convenient to relate this parameter to the variance of the conserved scalar. This relation has been established by Jiang et al. (1992):

$$I_s=\frac{\sigma^2(t)}{\sigma^2(0)}=\frac{2}{\pi}\arctan\left(\frac{2}{\gamma\sqrt{\gamma^2+4}}\right), \tag{4}$$

where I_s is known as the "intensity of segregation" (Brodkey, 1975). The parameter γ can also be related to the ensemble-mean value of the reacting scalar. This relation is obtained by substituting Eq. 3 into Eq. 1. After significant algebraic operations, the final results yield (Frankel, 1993):

$$\frac{\langle A\rangle(t)}{\langle A\rangle(0)}=1-\frac{2\arctan\left(\frac{\gamma}{\sqrt{2}}\right)}{\pi} \tag{5}$$

Now, with the establishment of Eqs. 4-5, the mean fractional conversion can be expressed in terms of I_s:

$$\mathcal{F}=1-\frac{\langle A\rangle(t)}{\langle A\rangle(0)}=1-\frac{2}{\pi}\arcsin\left(\sqrt{\sin\left[\frac{\pi I_s}{2}\right]}\right) \tag{6}$$

Assumed Frequencies

Miller et al. (1993) have recently shown that for the binary mixing problem considered here, the AMC can be viewed as a member of the general family of distributions generated by the Johnson-Edgeworth transformation (JET) (Johnson, 1949a; Edgeworth, 1907). With this transformation, alternative frequencies can be developed for the modeling of turbulent mixing. One such frequency that has proven effective is the "Logit-normal" distribution (Miller et al., 1993):

$$\mathcal{P}(\psi)=\frac{\Delta}{\sqrt{2\pi}\psi(1-\psi)}\exp\left\{-\left[\frac{\Delta^2}{2}\log^2\left(\frac{\psi}{1-\psi}\right)\right]\right\}, \tag{7}$$

where Δ, analogous to γ in the AMC, is not known *a priori*. For this PDF, neither the variance nor the reactant decay can be determined analytically, and their evaluation is possible solely by numerical means (Johnson, 1949a).

The similarity between the Logit-normal distribution and the family of Pearson (1895) frequencies suggests the use of the beta density of the first kind as a potential assumed PDF for the modeling of the mixing phenomena. For a random scalar variable within [0,1], this density is parameterized by its first two moments (Casella and Berger, 1990):

$$\mathcal{P}(\psi)=\psi^{\beta_1-1}(1-\psi)^{\beta_2-1}\frac{\Gamma(\beta_1+\beta_2)}{\Gamma(\beta_1)\Gamma(\beta_2)}, \tag{8}$$

where Γ is the Gamma function (Abramowitz and Stegun, 1972), and β_1 and β_2 are determined from the first two moments of the random variable. With a combination of this equation and Eq. 1, the mean fractional conversion can be obtained analytically. Following the same procedure as that for the AMC, the final result after some manipulations can be expressed as:

$$\mathcal{F}=1-\frac{\Gamma\left(\frac{1}{2I_s}\right)}{\sqrt{\pi}\Gamma\left(\frac{1}{2I_s}+\frac{1}{2}\right)} \tag{9}$$

Comparisons

The relations for the mean fractional conversion obtained by the models described above are compared with those obtained by employing the 3E model of Dutta and Tarbell (1989) and Tarbell (1992):

$$\mathcal{F}=\frac{1-I_s}{1+I_s}, \tag{10}$$

and that based on Toor's hypothesis (Toor, 1962, 1975):

$$\mathcal{F}=1-\sqrt{I_s}. \tag{11}$$

To examine the performance of the models, the predicted results via all the closures are compared against DNS data. The DNS procedure is similar to that of previous simulations of this type (Givi and McMurtry, 1988; Madnia and Givi, 1993). The subject of the present DNS is the three-dimensional periodic homogeneous box flow under the influence of a binary reaction of the type described above. The initial species field is assumed to be composed of out-of-phase square waves for the two reactants A and B. The computational package is based on the modification of a spectral-collocation procedure using Fourier basis functions developed by Erlebacher et al. (1987, 1990a, 1990b). The hydrodynamic field is assumed isotropic and is initialized in a similar manner to that of Erlebacher et al. (1990a). The turbulent field is of a decaying nature in that

there is no artificial forcing mechanism to feed energy to low wave numbers. The code is capable of simulating flows with different levels of compressibility (Erlebacher et al., 1990b; Hussaini et al., 1990). Here, only the results obtained for a low compressible case are discussed, since most previous analyses of plug-flow reactors have dealt primarily with incompressible flows (Toor, 1975; Hill, 1976; Brodkey, 1981; Leonard and Hill, 1991). The resolution consists of 96 collocation points in each direction. Therefore, at each time step 96^3 is the sample size for statistical analyses. This resolution allows simulations with a Reynolds number (based on the Taylor microscale) of $Re_\lambda \approx 41$. The value of the molecular Schmidt number is set equal to unity.

The PDFs generated by the AMC, JET, and the beta density are somewhat similar. For the binary mixing problem, all these models are capable of predicting the PDF evolution from an initially double delta state (segregated reactants) to an asymptotic Gaussian-like distribution near the mean at small variances. This behavior cannot be predicted by any of the other closures considered here.

Figure 1 shows the temporal variation of the fractional conversion for these three models along with the DNS data. All these closures yield almost identical results and exhibit good agreement with the DNS data during all stages of the mixing process. In light of this, to compare with the other closures, only the AMC is considered, as shown in Figure 2. At large times, the profiles are bounded above and below by the 3E model and the Toor closure, respectively. Initially, the results via the 3E closure are very close to those obtained by AMC. At all times, the results generated by the numerical integration of the C/D models of Curl (1963) and Janicka et al. (1979) overpredict the DNS data more than the simple expression generated by the AMC. Finally, as indicated before, the results obtained by LMSE are identical to those based on Toor's hypothesis. These comparisons indicate that the AMC, the beta density, and the Logit-Normal distribution yield the best overall behavior in predicting the rate of reactant conversion in accordance with the DNS data. This agreement follows from the compatibility of the model PDFs with those of the DNS, at least for the case of binary mixing considered here. Furthermore, a nice feature of these models is the explicit form of the final equations expressing these statistical quantities. It is noted that explicit analytical relations can be obtained only for the AMC and the beta density. Therefore, in the absence of better alternatives Eqs. 6 and 9 are recommended for effective and practical modeling of unpremixed plug-flow reactors.

Despite the favorable features of our simple mathematical expressions, the ramifications of the assumptions made in deriving these expressions must be emphasized. First, due to the assumption of infinitely fast chemistry, only the limiting rate of reactant conversion is obtained. The extensions to finite rate chemistry, reversible reactions, and multistep kinetics systems require numerical integration of the PDF. For these cases, the implementation of AMC for the binary case is straightforward, since it provides a closure for the joint PDFs of the scalar quantities (Pope, 1991). However, the validity of a multivariate beta (usually known as the "Dirichlet" (Johnson and Kotz, 1972; Wilks, 1962)) distribution and multivariate forms of the JET generated frequencies (Johnson, 1949b) cannot be guaranteed for general applications. Secondly, the simple formulas presented here are valid only for initially segregated reactants in stoichiometric proportion. This condition is compatible with the majority of previous works on nonpremixed plug-flow reactors (Toor, 1975; Brodkey, 1981; Leonard and Hill, 1988; Kosaly and Givi, 1987; Givi and McMurtry, 1988; Givi, 1989). The AMC and the beta density can be used for modeling nonstoichiometric mixtures, but the final results can be evaluated only by numerical means. For more complicated initial conditions, the use of beta and JET generated frequencies cannot be justified, while the AMC can be utilized in conjunction with appropriate numerical algorithms (Pope, 1991; Valiño et al., 1991). Finally, in the context of a single-point PDF formulation, there is no information pertaining to the evolution of the relevant turbulent length scales. Therefore, the final expressions can be only presented in terms of I_s or other related physical parameters (such as, σ^2, γ, and Δ). In this context, these parameters must be provided by external means, including experimental data and turbulence models (Frankel et al., 1992). Also, in nonequilibrium reacting systems, the segregation parameter should be defined to include

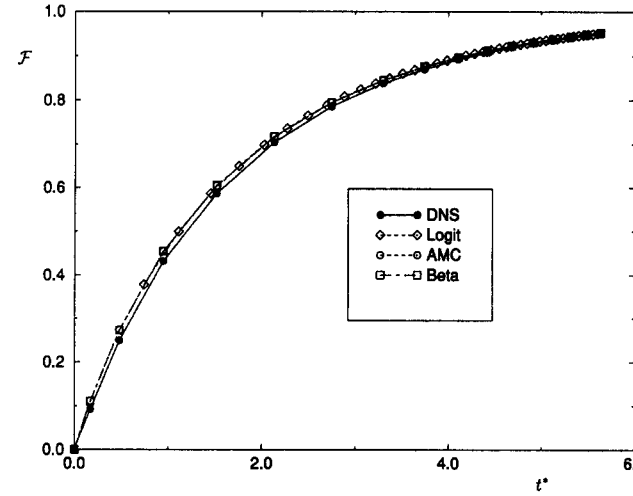

Figure 1. Fractional conversion vs. normalized time.

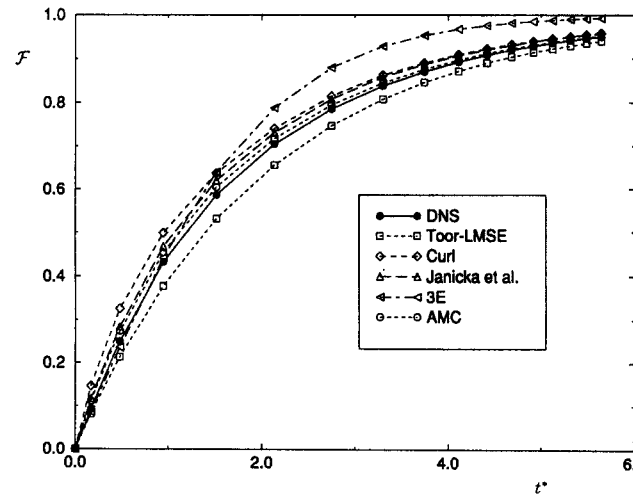

Figure 2. Fractional conversion vs. normalized time.

the information pertaining to the length-scale evolution of the reacting scalar.

In spite of these assumptions, it is very encouraging to have physically plausible algebraic relations for direct and accurate estimate of the reactant conversion rate in plug-flow reactors. Because of the simplicity of our final results, these expressions are recommended for routine and economical engineering predictions in nonpremixed binary reacting systems such as those in batch mixers.

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Notation

A, B = reactant concentration
$\mathfrak{A}$ = kernel of the C/D closure
I_s = intensity of segregation
$\mathfrak{I}$ = conserved scalar variable
$\mathfrak{F}$ = fractional conversion
t = time
t^* = normalized time $= -\ln[\sigma^2(t)/\sigma^2(0)]$

Greek letters

δ = Delta function
Δ = parameter of Logit-normal distribution
γ = parameter of the AMC closure
Γ = gamma function
σ^2 = variance of the conserved scalar variable
ω = frequency of mixing

Subscripts

0 = time zero (inlet of plug-flow reactor)

Other symbol

$\langle\ \rangle$ = ensemble average

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